

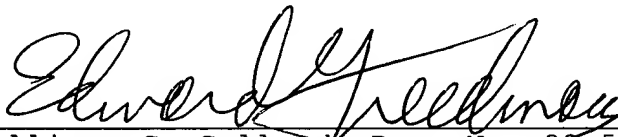
REMARKS

By this Preliminary Amendment, the application has been amended to conform with U.S. practice, the cross-reference to the related application has been inserted on page 1. Also, claims 1-13 have been replaced by new claims 14-26. No new matter has been introduced.

Entry of this amendment is respectfully requested.

Respectfully submitted,

FRIEDEMANN PIESCHEL ET AL



COLLARD & ROE, P.C.
1077 Northern Boulevard
Roslyn, New York 11576
(516) 365-9802

Allison C. Collard, Reg. No. 22,532
Edward R. Freedman, Reg. No. 26,048
Attorneys for Applicants

Express Mail No. EL 871 449 956 US

Date of Deposit November 28, 2001

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. § 1.10, on the date indicated above, and is addressed to U.S. Patent and Trademark Office, P.O. Box 2327, Arlington, VA 22202


Lisa L. Vulpis

capacity and are additionally capable of absorbing and binding undesirable foreign or attendant substances in the filter medium such as, for example hardening constituents or heavy metals. Furthermore, the problem of the invention is to provide suitable methods for producing the filters.

The problem is solved according to the invention by the features specified in claim 1. The features relating to the proposed methods for producing the filters are the objects of claims 2 to 13.

The filters made of filter paper or paper-like nonwoven material partially or wholly consist of fibrous material containing cellulose. The properties of the filters are substantially enhanced by a special treatment of the material containing cellulose, either prior to or after the manufacture of the filter paper. According to the invention, the treatment is carried out in such a manner that the material containing cellulose is at least partially carbamided with urea up to a nitrogen content of 1 to 4% by mass bonded in amino-methane acid ester groups (carbamide groups), and phosphorylated with phosphoric acid or ammonium phosphate up to a phosphorus content of 3 to 8% by mass. In addition to high filtration capacity, the filter produced from cellulose-containing material so modified additionally possess the special properties of binding hardening constituents as well as toxic heavy metals, which impair the

within the specified range limits depending on the purpose of application of the filters.

The phosphorylation and carbamidation of the cellulose-containing starting material for the production of the filter paper or the paper-type nonwoven material is carried out under the following conditions:

It is important that the cellulose-containing fiber material is brought into a particularly reactive form prior to the phosphorylation and carbamidation reaction. Such a so-called activation is carried out by adjusting the moisture content of the cellulose-containing ^{*}[fibers to a value in excess of 30% in particular by adding water]. The cellulose-containing starting material usually already has a water content of from 5 to 25%. In order to achieve the desired activation it is necessary that the cellulose-containing fiber material is subjected to the action of water over a longer period of time. The duration is substantially dependent upon the already existing moisture content of the material and amounts to at least half an hour.

The reaction partners phosphoric acid or ammonium phosphate and urea have to be admixed to the cellulose-containing material in such a way that said reaction partners are present in the material with uniform distribution after the mixing process has been completed. In * material by adding water to it in an amount of at least 30% by mass of the cellulose-containing material

phosphorylation and carbamidation reaction, -which is carried out [under vacuum as well]. by heating the mixture to a temperature of 125 to 155°C while simultaneously applying a vacuum and maintaining a reaction time of at least 15 minutes.

Carrying out said reaction under vacuum leads to a number of decisive advantages. Of great importance is that the reaction temperature can be reduced by about 40°C as compared to when it is carried out under normal pressure. Secondary reactions of phosphoric acid or ammonium phosphate and urea are distinctly reduced in this way, and decomposition reactions of the cellulose-containing fibrous material are suppressed. For example, it is possible, furthermore, to reduce the amounts of the reaction components urea and phosphoric acid or ammonium phosphate used. Furthermore, a careful treatment of the cellulose-containing material is assured as the phosphorylation and carbamidation is being carried out owing to the low reaction temperatures and reduced amounts of phosphoric acid or ammonium phosphate and urea used. In this way, the structures and mechanical properties of the cellulose-containing fibrous materials are preserved in the course of the reaction to a large extent, which is very important for the manufacture of the paper or nonwoven material.

Furthermore, it is important to maintain reaction times of at least 15 minutes. If the reaction times are shorter, the phosphoric acid used, for example, is reacted incompletely, and in particular the nitrogen content will be

starting material is treated with a solution of phosphoric acid and/or ammonium phosphate ^{and urea} in water at a molar ratio of urea to phosphorus of 2.5 : 1 to 4.5 : 1, whereby the amount of water is adjusted in such a way that 1 to 8 mols phosphorus per kg cellulose remain in the cellulose-containing starting material. The starting material can be treated on one or both sides by coating it with the solution, or it is impregnated in a bath of the solution in a device operating in cycles.

The water is completely expelled by a subsequent vacuum treatment with simultaneous heating of the starting material to a temperature of 60° to 100°C. Thereafter, the phosphorylation and carbamidation reaction is carried out under vacuum as well, at a temperature of 125° to 155°C and

in the course of a reaction time of at least 15 minutes. The vacuum is preferably adjusted in each case to a value of 5.33 kPa to 26.66 kPa

The phosphorylated and carbamided starting material is subsequently cooled, washed phosphate-free, and finally dried. The desired filters are then produced from the modified filter paper or nonwoven material in the manner known per se by punching, folding and winding.

In connection with filters used in applications for drinking water, the present ammonium form is converted before the phosphorylated and carbamided cellulose-containing material is washed and dried into the sodium form by treating it with a solution of common salt. The treatment

is carried out either on modified fibers prior to the actual manufacture of the paper, or on the modified filter paper or nonwoven material.

Example 1

100 g cotton linters (linters 503 of the Buckeye Mephis Company) present in the form of cardboard-like webs was cut into pieces. In a dish, a solution prepared at 60°C from 74.7 ml water, 61.4 g 85% phosphoric acid and 111.3 g urea was poured over said pieces and the dish was turned over frequently. After the solution was completely and uniformly absorbed, the dish was covered airtight and stored for one hour at room temperature. The dish was subsequently placed in a vacuum drying cabinet, a vacuum of 5.33 kPa ~~40 Torr~~ was applied, and drying was carried out at 90° to 100°C. When no more steam was left to be removed by suction, the temperature was raised to 140°C and maintained for 1.5 hours, whereby the vacuum was maintained as well. Obtained was 191.8 g of an externally unchanged reaction product, which was stirred into water, filtered off and washed until the wash water was free of phosphate. The product was dried in the drying cabinet at 110°C, whereby the yield came to 149.3 g.

A sample of the fiber material so obtained was converted by washing with concentrated common salt solution from the ammonium form into the sodium form, washed free of the salt,

and subsequently dried. The elementary analysis of said specimen resulted in a phosphorus content of 5.6% ^{by mass} and a nitrogen content of 1.3% by mass.

The fiber material so prepared was subsequently tested for its sorptive properties.

The sorption equilibrium data were determined according to the following method:

250 ml measuring flasks were loaded with the fiber samples (0.1 to 0.025 g) and each charged with 1 to 5 ml m/10 solutions of salts of the metals Cu and Ca, filled up, provided with magnetic stirrers, and stirred for 3 hours at room temperature. Upon settlement, the solutions were decanted, their pH was determined, and the metal content was determined complexometrically. The equilibrium concentrations in the fiber were calculated based on the equilibrium concentrations in the solution so obtained and on the starting concentrations fixed by the addition of metal salt solutions. By adding corresponding amounts of nitric acid before the measuring flasks were filled up, the pH in the sorption was adjusted to pH = 4.5. Several control measurements of the equilibrium concentrations in the solution by means of atom absorption spectroscopy (AAS) showed deviations in the range of the measuring accuracy and in this way confirmed the reliability of complexo-metric analyses in the sorption tests.

The result was a filtrate with 0.4 °dH and a copper content of 0.01 mg/l.

Example 2

100 g filter paper consisting of spruce cellulose for laboratory purposes, which was present in the form of sheets in the DIN A4-format, was placed on a substrate and uniformly coated with a solution of 28.3 g ammonium phosphate and 50.9 g urea in 126 ml water, whereby the entire amount of the solution was consumed. After 30 minutes, the substrates with the sheets were place in a vacuum drying cabinet, a vacuum of $\frac{6.67 \text{ kPa}}{50 \text{ Torr}}$ was applied, and all water was expelled by heating to 100°C. The temperature was raised within 30 minutes to 155°C. This temperature was maintained for 30 minutes, and venting and cooling was then carried out rapidly. The result was 137.7 g product, which could be washed free of phosphate by careful washing while preserving the original shape of the sheets. After the sheets were dried at 110°C in the normal drying cabinet, 121.0 g treated filter paper was obtained as the result.

The elementary analysis following conversion into the Na-form as in example 1 showed a phosphorus content of 3.3%^{by mass} and a nitrogen content of 1.9%by mass.

The sorption capacity determined analogous to example 1 showed for copper 66.7 mg Cu/g filter, and for calcium 44.1